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DATE: December 3, 2001

FORM PTO 1390 (REV 11-2000) U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		CENTRO-109
		US APPLICATION NO (If known, see 37 CFR 1 5) 09/980936
INTERNATIONAL APPLICATION NO. PCT/EP00/05128	INTERNATIONAL FILING DATES 05 June 2000	PRIORITY DATE CLAIMED 04 June 1999
TITLE OF INVENTION FUNCTIONALIZED THERMOPLASTIC POLYMER COMPOSITION AND A PROCESS FOR ITS PREPARATION		
APPLICANT(S) FOR DO/EO/US Francesco Mascia and Franca Arrighi		

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [x] This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. [ ] This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing 35 U.S.C 371
3. [x] This is an express request to begin national examination procedures (35 U.S.C. 371 (f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. [x] The US has been elected by the expiration of 19 months from the priority date (PCT Article 31).
5. [x] A copy of the International Application as filed (35 U.S.C. 371 (c)(2))
   - a [ ] is attached hereto (required only if not communicated by the International Bureau).
   - b. [x] has been communicated by the International Bureau.
   - c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [ ] An English language translation of the International Application as filed (35 U.S.C. 371 (c)(2)).
   - a. [ ] is attached hereto.
   - b [ ] has been previously submitted under 35 U.S.C. 154(d)(4).
7. [x] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   - a. [ ] are attached hereto (required only if not communicated by the International Bureau).
   - b. [ ] have been communicated by the International Bureau.
   - c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   - d. [x] have not been made and will not be made.
8. [ ] An English language translation of the amendments to the claims under PCT Article 19 (35 U S.C. 371 (c)(3)).
9. [x] An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). (Unexecuted)
10. [ ] An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S C. 371 (c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. [x] An Information Disclosure Statement under 37 CFR 1.97 and 1.98 w/PTO-1449, 3 references
12. [ ] An assignment document for recording A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. [ ] A FIRST preliminary amendment.
14. [ ] A SECOND or SUBSEQUENT preliminary amendment.
15. [ ] A substitute specification.
16. [ ] A change of power of attorney and/or address letter
17. [ ] A computer-readable form of the sequence listing in accordance with PCT Rule 13ter 2 and 35 U.S.C. 1 821 - 1.825.
18. [x] A second copy of the published international application under 35 U S.C 154(d)(4).
19. [ ] A second copy of the English language translation of the international application under 35 U S.C 154(d)(4).
20. [x] Other items or information: Copy of International Preliminary Examination Report

EXPRESS MAIL LABEL NO.: EL651341481US
DATE: December 3, 2001

U.S. APPLICATION NO (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO	ATTORNEY'S DOCKET NUMBER
09/980936	PCT/EP00/05128	CENTRO-109

17. [x] The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) – (5)):

		CALCULATIONS PTO USE ONLY
[ ]	Neither international preliminary examination fee (37 CFR 1 482) nor international search fee (37 CFR 1 445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO	$1040 00
[x]	International preliminary examination fee (37 CFR 1 482) not paid to USPTO but International Search Report prepared by the EPO or JPO	$890 00
[ ]	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO . .	$740 00
[ ]	International preliminary examination fee (37 CFR 1 482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)	$710.00
[ ]	International preliminary examination fee (37 CFR 1 482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)	$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 890.00	
Surcharge of $____ for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	$	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	16-20 =		x	$ 0.00	
Independent claims	4-3 =	1	x 84.00	$ 84.00	
MULTIPLE DEPENDENT CLAIM(s) (if applicable)			+ 280.00	$ 280.00	
TOTAL OF ABOVE CALCULATIONS =				$ 1,254.00	
[ ] Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by ½				$	
SUBTOTAL =				$ 1,254.00	
Processing fee of $____ for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492 (f)). +				$	
TOTAL NATIONAL FEE =				$ 1,254.00	
Fee for recording the enclosed assignment (37 CFR 1 21 (h)). Assignment must be accompanied by appropriate cover sheet (37 CFR 3.28, 3.31) + (per property).				$	
TOTAL FEES ENCLOSED =				$ 1,254.00	
				Amount to be Refunded:	$
				Charged:	$

a. [ ] A check in the amount of $ ____ to cover the above fees is enclosed.

b. [x] Please charge my Deposit Account No. 12-1095 in the amount of $ 1,254.00 to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [x] The Commissioner is hereby authorized to charge any additional fees which may be required or credit any overpayment to my Deposit Account No. 12-1095. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO

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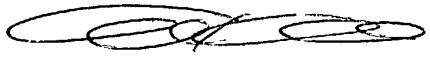

SIGNATURE

Arnold H Krumholz

NAME

25,428

REGISTRATION NUMBER

"A FUNCTIONALIZED THERMOPLASTIC POLYMER COMPOSITION AND A PROCESS FOR ITS PREPARATION"

Technical Field

The present invention relates to a polyolefin-based functionalized thermoplastic polymer formulation which can be injection-molded, has a good hand (soft-touch), a high scratch-resistance, and is suitable to various forms of finishing, such as welding, gluing and painting. Particularly, the polymer formulation according to the invention is suitable to such uses as the external material - or skin - of trim panels for autovehicles.

The invention relates further to a process for the preparation of the aforementioned formulation.

Background Art

Various types of injection-moldable polyolefin-based polymers with a soft-touch are known in the art.

Application WO96/22327 describes a polymer formulation suitable for injection molding comprising 5-65% by weight of EP(D)M, 1-30% of polyalkanylene and their mixtures, grafted onto a matrix of polypropylene. This formulation has good processability and reduced stickyness, but, like other similar formulations, has little scratch-resistance and can not be subjected to welding, gluing or painting processes.

Another problem of known formulations is their processability.

Like the above mentioned ones, these materials are used to produce the external layer of skin in such products as car trim panels; the area of the panel is relatively wide while the thickness of the skin layer is very reduced. The material used should therefore have excellent

processability to be able to distribute itself in the die avoiding the form of "tiger-stripes" (advancement fronts) i.e. the stripes or signs due to the different rheological behavior of the polyolefin matrix with respect to the rubbery part.

The known formulations do not satisfy the above mentioned requirements and produce products having a high number of tiger stripes.

There is a need, therefore, to have formulations with the aforementioned improved features.

Disclosure of the invention

Such purpose is achieved by the present invention which relates to a polyolefin-based thermoplastic polymer formulation characterized by comprising, by percentage of the total polymer weight, 10-80% of polypropylene and, grafted thereon, 10-85% of EP(D)M rubber, 0-40% of polybutadiene, and 0.5-60% of at least one unsaturated compound selected from: ethylene-vinyl acetate copolymers and terpolymers, NBR, acrylic ethylene-ester copolymers and terpolymers and polybutadiene polyurethanes or their precursors, the total amount of polybutadiene and unsaturated compounds not exceeding 60% by weight.

According to a preferential embodiment of the invention, the formulation contains polybutadiene-based polyurethanes obtained from precursors consisting of functinalized polybutadienes, i.e polybutadienes having -OH, -NCO, -COOH groups.

According to a further embodiment the invention, the formulation contains the above mentioned precursory functionalized polybutadienes instead of or in addition to the polyurethane

obtained therefrom.

According to another preferential aspect, the formulation also comprises 5-30% of cellulose by weight of the total of the formulation.

The invention further relates to a composition for the preparation of a polymer formulation of the type above described, characterized according to Claim 5.

The invention further relates to a process for the production of a thermoplastic polymer formulation, characterized according to Claim 8.

The invention further relates to a trim element for vehicles, provided with a surface at least partly made with a material whose formulation is of the type described above. In a preferential embodiment, the product has an internal support layer and an external skin layer produced with the formulation of the invention.

The formulation according to the present invention has numerous advantages. The layer of "skin" obtained with the formulation of the invention is provided with particularly high scratch-resistance values, while also having excellent soft-touch and good mechanical characteristics.

The formulation presents excellent processability that makes it perfectly suitable both for being extruded in a flat layer, and for injection molding, and particularly for co-injection molding of products in which the formulation according to the invention constitutes the external layer or "skin."

A further advantage is that the products obtained from a material having the formulation of the invention can be subjected to finishing processes and particularly to painting, glueing and welding, e.g. high

frequency welding.

The thermoplastic polymer formulation according to the invention comprises, by percentage of the total weight of polymers (i.e. excluding the weight of possible fillers), 10-80% of polypropylene, 0-40% of polybutadiene, 10-85% by weight of EP(D)M and 0,5-60% of an unsaturated compound selected from among ethylene-vinyl acetate copolymers and terpolymers (e.g. EVA), NBR (nitrile-butadiene rubber), acrylic ethylene-ester copolymers and terpolymers (e.g. EMA, EBA), and polybutadiene-polyurethanes or their precursors.

The total amount of unsaturated compounds (that is EVA etc., NBR, EMA, EBA etc. and polybutadiene-polyurethane) and of polybutadiene is less than or equal to 60% by weight of the polymers. The EP(D)M rubber and the polybutadiene and polybutadiene containing compounds are partially cross-linked and grafted onto the polypropylene; namely, the unsaturated compounds act as grafting units.

Precursors of the polybutadiene-polyurethanes are here meaning functionalised polybutadienes with terminal groups selected from -NCO, -OH and -COOH.

Polypropylenes suitable for use in the invention are propylene polymers and copolymers with isotatic index greater than 30%, such as the copolymers of propylene with one or more alpha-olefines having 2-10 atoms of carbon, as well as formulations of polypropylene obtained with sequential polymerization of polypropylene and of its mixtures with ethylene and/or alpha-olefines. Isotactic (co)polymers are preferred. The suitable propylene (co)polymers have a MFI (Melt Flow Index) that can vary over a wide range and is preferably within

the range of 0.1 and 60 at 230°C/ 21.6 N (ASTM 1238). Examples of suitable polypropylenes are those distributed commercially by Borealis under the proprietary name DAPLEN® and the codes DS10, US105A, CHC3007, FSC1012, and USC 1012.

The amount of polypropylene present is preferably between 15 and 50%.

EP(D)M rubbers (including EPR) suitable for use in the present invention are EP(D)M polymers and rubbers and their analogs; they are known in the art and are available commercially. Examples of suitable EP(D)M polymers are those commercially available from Enichem Elastomeri under the name of DUTRAL®; similar known examples of EP(D)M polymers are random copolymers having amorphous content greater than 50% by weight, for instance C_2–C_8 type copolymers such as ENGAGE®, from Du Pont Dow Elastomers. The EP(D)M rubbers are, by preference, oil extended. Suitable oils for the extension are known in the art and commercially available, such as naftene and paraffin oils for instance. The amount of oil is generally between 40 and 60% by weight of the rubber and preferably around 50% by weight.

Preferably the amount of EP(D)M rubber present is 40-60% if an oil-extended rubber is used or 20-50% if dry.

Polybutadiene suitable to be used in the invention, is e.g. liquid polybutadiene containing at least 20% of structure 1,2, such as the one known as Lithene PH® of Revertex Ltd. Solid polybutadienes can also be used; this type of polybutadiene has a lesser content of 1,2 structure (usually 0.5 to 12%) and is commercially available, e.g. from Enichem Elastomeri with the trade names Intene and Europrene. Solid polybutadiene being a rubbery material, its use will result in a lesser

amount of EP(D)M/EPR rubbers in the final formulation; e.g. if Intene or a similar compound is used the amount of EP(D)M rubber will decrease from 40-60% to 20-50%.

Preferably, the amount of polybutadiene is between 0.5% and 10%; when polyurethanes (TPU) obtained from polybutadienes as below disclosed are used in the formulation, unfunctionalized polybutadiene (i.e. the previously disclosed polybutadienes such as Lithene or Europrene) is preferably not present or is present in a reduced amount (up to 1.0%).

Suitable ethylene-vinyl acetate copolymers and terpolymers are those which contain between 6% and 26% of vinyl acetate by weight and preferably between 9% and 18% by weight. An example of ethylene-vinyl acetate copolymers is EVA, for instance that known as Escorene FL909, from Exxon. Suitable terpolymers are ethylene-vinyl acetate-maleic-anhydride, for instance that commercialized under the name of OREVAC by ELF ATOCHEM. Preferably the amount of EVA and terpolymers is between 10% and 25% and none of these polymers is present when polybutadiene-polyurethanes are provided in the formulation.

Suitable NBR polymers are butadiene-acrylonitrile copolymers with acrylonitrile content within the range 25-35%. An example of suitable NBR polymer is the one known as CHEMIGUM P86F from Goodyear, containing between 30.5 and 33,5% of acrylonitrile.

Suitable acrylic ethylene-ester copolymers are random ethylene-acrylic-ester copolymers such as EMA (ethylene-methyl acrylate) and EBA (ethylene-butyl acrylate) containing between 6 and 22% by weight of acrylate. Copolymers of this type are commercialized under

the LOTRYL name by ELF ATOCHEM. Other suitable acrylate polymers are ethylene-ester acrylic-maleic- anhydride terpolymers (with acrylate content between 6-22% by weight) commercialized under the name of LOTADER by ELF ATOCHEM.

Suitable polyurethane polymers are those unsaturated, based on polybutadiene, i.e. polymers obtained by reacting polybutadiene functionalized with isocyanide, hydroxyl or acidic terminal groups; examples of these prepolymers are reported below.

According to the invention, besides the polybutadiene-based-polyurethanes, their precursors may also be used, i.e. polybutadienes prepared with functional terminal groups selected from -OH, -NCO, -COOH. Such precursors can also be used alone, instead of the other mentioned unsaturated compounds. Preferably,

It is believed that these functionalized polybutadienes, like the other unsaturated compounds (polybutadiene-polyurethanes, EVA, NBR), behave in a way analogous to polybutadiene, i.e. that they partly cross-link and become grafted onto the polypropylene and the EP(D)M rubbers during the process of production of the formulation by means of vis breaking. In this way, polymers that are incompatible (polypropylene and polyurethane) are made compatible and a polymer formulation - i.e. a material - is obtained provided with polar groups, that make the material suitable to be painted, welded or glued. Preferably, the amount of polybutadiene-based polyurethane or its precursors is between 8% and 45%, the amount of rubber is 30-55% and the amount of polypropylene is 30-65%.

The total amount of unsaturated compounds, i.e. polybutadiene, EVA, EVA terpolymers, NBR, ethylene-acrylic-ester copolymers and

terpolymers, polyurethanes and precursors (polybutadienes with functional groups), is equal to a maximum of 60% by weight.

In an embodiment of the invention the composition that must be subjected to reactive extrusion (vis-breaking) comprises both polybutadiene functionalized with hydroxyl groups and polybutadiene functionalized with isocyanide groups. It is believed that the polybutadiene prepolymers react together during the visbreaking reaction to give a polyurethane grafted onto the polypropylene.

In another embodiment, the polybutadiene-based polyurethane to be used is first produced starting from mixtures of the following compounds: di-isocyanates, including polybutadiene functionalized with -NCO terminal groups, diphenylmethane di-isocyanate (MDI), toluene di-isocyanate (TDI) and diols such as butanediol, hexanediol, polybutadiene functionalized with hydroxyl terminal groups.

Polyurethane precursors of the type described above, suitable for the present invention are for instance those commercially available under the name KRASOL®, LBD and LBH, from KAUCUK (Czech Republic).

An example of the synthesis of a polybutadiene-based polyurethane starting from a polybutadiene isocyanate and two types of diol, a hexanediol and a hydroxylated polybutadiene, is reported in the following examples. It was found that polyurethanes obtained from polybutadiene isocyanate (a polybutadiene polyol carrying two TDI end groups) and polybutadiene polyol and/or a known diol (e.g. a short-chain diol) are perfectly mixable with the rubber and the polypropylene and have good mechanical properties.

A preferential embodiment of the invention provides for the presence

of 5-30% (by weight of the total formulation) of cellulose. Cellulose suitable for to the purpose is the pure or substantially pure one such as, for instance, that commercially available under the name ARBOCELL ZZ600 and BEM 600 from JRS (Germany).

Preferably, from 10% to 20 % of cellulose is used. It has been surprisingly found that the presence of cellulose results in a remarkable increase in the scratch-resistance of the product and in an improvement of the mechanical properties, without this negatively influencing the soft-touch features of the product.

The invention further protects the compositions for preparing a formulation of the type described above. The term formulation means the material, for instance in granules, powder or pellets, into which rubbers and unsaturated compounds have already been converted by reactive extrusion (vis-breaking) and presumably are partially cross-linked and grafted onto the polypropylene. The material of the formulation is ready to be molded and is already endowed with the desired characteristics.

The term composition means the mixture of polymers and additives before their processing by means of reactive extrusion (vis-breaking) to give the formulation of the invention.

According to the present invention the composition comprises, in an analogous way to that described above, as a percentage by weight of the total weight of the polymers, 10-80% of polypropylene, 10-85% of an EP(D)M rubber, 0-40% of polybutadiene, and 0,5-60% of at least one unsaturated compound selected from ethylene-vinyl acetate copolymers and terpolymers, NBR, ethylene-acrylic-ester copolymers and terpolymers, and polybutadiene-based polyurethanes or their

precursors, where the total amount of unsaturated compounds and polybutadiene is less than or equal to 60% by weight.

Beyond these components, the composition further comprises 0.1-1.5% of a radical-generating agent at the processing temperature of the composition, that is when the composition is subjected to vis-breaking. The suitable polymers are those listed above.

Preferential agents able to generate radicals at the processing temperature of the composition are peroxides, such as, for instance, dicumyl peroxide, and 2,5-bis(tert-butylperoxyl)2,5-dimethyl hexane. Other suitable agents are non-peroxide initiators such as 2,3-dimethyl-2,3-diphenyl hexane and 2,3-dimethyl-2,3-diphenil butane. All the radicals-generating agents are able to generate radicals, and therefore to start cross-linking, at the processing temperature of the composition, i.e. at the temperature reached by the composition when it has been melted, kneaded and is then extruded.

The process for the production of a thermoplastic polymer formulation according to the invention provides for the kneading in the molten state of a composition of the type described above, i.e. containing the radical-generating agents, at such a temperature as to activate said radical generating agents to create a plurality of active sites on the polypropylene chain. At least partial cross-linking of the rubbers and a greater or lesser degradation of the polypropylene is obtained this way. In this phase, known also as "visbreaking" or "reactive extrusion", the polybutadiene acts as a grafting agent and grafts onto the polypropylene and the rubbers. It is presumed that, similarly, unsaturated compounds EVA and ethylene-vinyl acetate-maleic anhydride terpolymers, NBR, ethylene-acrylic ester co- and

terpolymers and polybutadiene-based polyurethanes and their precursors also behave as grafting units on the polypropylene and the rubbers. The resultant composition will give a material with excellent mechanical and soft-touch characteristics and high scratch-resistance.

The temperature at which this process of reactive extrusion takes place is generally between 200 and 220 °C.

In order to avoid thermo-degradation phenomena of the components of the mixture, particularly during the reactive extrusion described above, antioxidants and stabilizers will be present in the usual amount, for instance between 0.1 and 1.5% by weight on the total weight of the polymers. These additives are known in the art; examples of suitable additives are sterically hindered phenols, secondary amines, thioethers, phosphites and phosphonites.

Preferably the amount of filler in the formulation is between 5-30%. Besides the cellulose described above, other vegetable fillers can be used, as well as additives and fillers such as for instance carbonate, silica and silicates like wollastonite, talc, caolin and dyes.

Best modes for carrying out the invention

The invention will be further illustrated with reference to the following examples.

Example A- Synthesis of the polybutadiene based polyurethane

175.2 parts of KRASOL LBD, a polybutadiene with isocyanate end groups from KAUCUK (Czech Rep.), 22.55 parts of KRASOL LBH 3000, a polybutadiene with hydroxyl end groups and 2.25 parts of 2-ethyl-1,3-hexanediol are mixed and reacted at 80°C for 48 hours. A solid is obtained which is milled to be used in example 4.

Example 1

44.8 parts of DUTRAL CO 555 (an EPM (EPR) polymer oil-extended 45%), 35.8 parts of DAPLEN FSC 1012 Polypropylene (MFI 5 at 230°C/ 5Kg), 17 parts of EVA ESCORENE-FL 909 (9.4% vinyl acetate content) from EXXON, 0.4 parts of a peroxide radical-generating agent di(t-butyl)peroxide-di-isopropylbenzene 40% on inactive support, 0.9 parts of antioxidant additives and 1.1 parts of LITHENE PH polybutadiene from REVERTEX CHEMICALS were mixed at 180-220°C in a Maris Ø 133 L/D= 40 co-rotating twin-screw extruder and then pelletized.

Example 2

48 parts of DUTRAL CS 9615 (an EPDM polymer oil-extended 45 %), 37.4 parts of DAPLEN FSC 1012 Polypropylene (MFI 5 at 230°C/ 5Kg), 10 parts of CHEMIGUM P86F from GOODYEAR (a butadiene-acrylonitrile rubber), 0.4 parts of peroxide radical-generating agent di(t-butyl) peroxide-di-isopropylbenzene 40 % on inactive support, 0.9 parts of antioxidant additives and

1.3 parts of LITHENE PH polybutadiene from REVERTEX CHEMICALS were mixed at 180-220°C in a Maris Ø 133 L/D= 40 co-rotating twin-screw extruder and then pelletized.

Example 3

52 parts of DUTRAL CS 9615 (an EPDM polymer oil-extended 45 %), 42 parts of Polypropylene DAPLEN FSC 1012 (MFI 5 at 230°C/ 5Kg), 0.4 parts of peroxide radical-generating agent di(t-butil) peroxide-di-isopropylbenzene at 40 % on inactive support, 0.4 parts of antioxidant additives, 1 part of LITHENE PH polybutadiene from REVERTEX CHEMICALS, 4 parts of KRASOL LBD (a polybutadiene with isocyanate end groups) and 0.2 parts of 2-etil-1,3-hexanediol (Aldrich)

were mixed at 180-220°C in a Maris Ø 133 L/D= 40 co-rotating twin-screw extruder and then pelletized.

Example 4

48 parts of DUTRAL CS 9615 (an EPDM polymer 45% oil-extended), 40 parts of Polypropylene DAPLEN FSC 1012 (MFI 5 at 230°C/ 5Kg), 10 parts of polybutadiene-based Polyurethane synthesized as described above, 0.4 parts of peroxide radical-generating agent di(t-butyl) peroxide-di-isopropylbenzene at 40% on inactive support, 0.7 parts of antioxidant additives and 0.9 parts of polybutadiene LITHENE PH of the REVERTEX CHEMICALS

were mixed at 180-220°C in a Maris Ø 133 L/D= 40 co-rotating twin-screw extruder and then pelletized.

The following table reports the values of some mechanical characteristics of the formulations obtained.

Table 1

	Example 1	Example 2	Example 3	Example 4
MFI at 230°/21 6N	8.8	5.5	14.6	9.5
Shore D	32	25	25	23

The materials obtained were molded by means of co-injection molding technique on 1850-ton Mir press at a temperature of about 220°C, to give products in which the formulation constitutes the external layer of skin.

The materials have good processability and distribution of the materials, as well as reduced fogging and improved scratch-resistance.

Further, it was observed a substantial reduction of the advancement

fronts (tiger-stripes) usually due to the different rheological behavior of the polyolefin matrix with respect to the rubber portion.

The polymer formulation as obtained is particularly suitable for the production of the skin of trim elements for cars where by the term "components " are understood all the known components for cars, such as panels, bridges, console, and coverings generally.

CLAIMS

1. A polyolefin-based thermoplastic polymer formulation characterized by comprising, by weight of the total weight of the polymers, 10-80% of polypropylene, 10-85% by weight of EP(D)M rubber, 0-40% of polybutadiene, and 0,5-60% of at least one unsaturated compound selected from: ethylene-vinyl acetate copolymers and terpolymers, NBR, ethylene-acrylic ester copolymers and terpolymers, and polybutadiene-based polyurethanes or their precursors, or mixtures thereof, the maximum total amount of polybutadiene and unsaturated compounds being 60% by weight, said polybutadiene and unsaturated compounds being grafted onto the polypropylene and the EP(D)M rubber.

2. A formulation according to Claim 1, in which said precursors of polybutadiene-based polyurethanes are one or more polybutadienes provided with terminal groups selected from -NCO, -OH, -COOH.

3. A formulation according to Claim 1 or 2, in which said EP(D)M rubbers are oil extended at 30-60% by weight.

4. A formulation according to any previous Claim, characterized by the polybutadiene-based polyurethane being the reaction product of a polybutadiene derivative having an -NCO group with a polybutadiene derivative having a terminal group selected from -OH and -COOH and/or with a diol known in the art.

5. A composition for the preparation of a formulation according to any preceding Claim, characterized by comprising by weight out of the total weight of the polymers, 10-80% of a polyolefin selected from polyethylene and polypropylene, 10-85% by weight of an EP(D)M rubber, 0-40% of a polybutadiene, 0,5-60% of at least one unsaturated

compound selected from ethylene-vinyl acetate copolymers and terpolymers, NBR, ethylene-acrylic ester copolymers and terpolymers, and polybutadiene-based polyurethanes or their precursors the maximum total amount of polybutadiene and of unsaturated compounds being 60% by weight.

6. A composition according to Claim 5, comprising 0,5-20% of one or more polybutadienes provided with terminal groups selected from :

-NCO, -OH,-COOH.

7. A composition according to claim 5 or 6, further comprising 0.1-1.5% of a radical-generating agent at the processing temperature of the composition.

8. A process for the production of a thermoplastic polymer formulation, characterized by mixing a composition comprising, by weight of the total weight of the polymers, 10-80% of a polyolefin selected from polyethylene and polypropylene, 10-85% by weight of an EP(D)M rubber, 0-40% of a polybutadiene, 0,5-60% of at least one unsaturated compound selected from ethylene-vinyl acetate copolymers and terpolymers, NBR, ethylene-acrylic ester copolymers and terpolymers, and polybutadiene-based polyurethanes or their precursors, the maximum total amount of polybutadiene and of unsaturated compounds being 60% by weight, and 0,1-0,6% of at least one agent generating radicals, at a temperature such as to activate said radical-generating agent to effect a partial cross-linking of at least some of the polymers present.

9. A process according to Claim 8, in which at least one polybutadiene provided with terminal groups selected from -OH,

-NCO and -COOH, is used as unsaturated compound.

10. A process according to Claim 8 or 9, in which said polybutadiene-based polyurethane is prepared by reaction of -NCO functionalized polybutadienes with a –OH or –COOH polybutadienes and/or a diol.

11. A process for the preparation of an unsaturated polybutadiene-based polyurethane, in which a polybutadiene prepolimer provided with isocyanate end groups is reacted with one or more diols.

12. A process for the preparation of an unsaturated polybutadiene-based polyurethane, in which a polybutadiene prepolimer provided with hydroxyl terminal groups is reacted with one or more di-isocyanates.

13. A trim element for autovehicles, characterized by having an external surface made at least partly with a thermoplastic material having a formulation according to any Claim 1 to 4.

14. A trim element for autovehicles according to Claim 13, comprising an internal support layer and an external skin layer, said external skin layer corresponding to said surface.

10 Rec'd PCT/PTO 30 MAY 2002

PTO/SB/01 (03-01)

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63)

[x] Declaration Submitted with Initial Filing OR [ ] Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required)

Attorney Docket Number	CENTRO-109
First Named Inventor	Francesco Mascia
COMPLETE IF KNOWN	
Application Number	Not Yet Assigned
Filing Date	
Group Art Unit	N/A
Examiner Name	Not Yet Assigned

As a below named inventor, I hereby declare that:

My residence, mailing address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

FUNCTIONALIZED THERMOPLASTIC POLYMER COMPOSITION AND A PROCESS FOR ITS PREPARATION

(Title of the invention)

the specification of which

[ ] is attached hereto

OR

[x] was filed on (MM/DD/YYYY) 06/05/2000 as United States Application Number or PCT International Application No. PCT/EP00/05128 and was amended on (MM/DD/YYYY) ______ (If applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56, including for continuation-in-part applications, material information which became available between the filing date of the prior application and the national or PCT international filing date of the continuation-in-part application.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or (f), or 365(b) of any foreign application(s) for patent, inventor's or plant breeder's rights certificate(s), or 365 (a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent, inventor's or plant breeder's rights certificate(s), or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	NO
99830351.5	EP	06/04/1999	[ ]	[ ]	[x]
			[ ]	[ ]	[ ]
			[ ]	[ ]	[ ]
			[ ]	[ ]	[ ]

[ ] Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

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(81) Designated States *(national)*: BR, JP, KR, US.

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 00/75225 A2

(54) Title: A FUNCTIONALIZED THERMOPLASTIC POLYMER COMPOSITION AND A PROCESS FOR ITS PREPARATION

(57) Abstract: A polyolefin-based thermoplastic formulation giving products with soft-touch and functionalized to be welded and painted comprises, by weight of the total weight of the polymers, 10-80 % of polypropylene, 10-85 % by weight of EP(D)M rubber, 0-40 % of polybutadiene, and 0.5-60 % of at least one unsaturated compound selected from: ethylene-vinyl acetate copolymers and terpolymers, NBR, ethylene-acrylic ester copolymers and terpolymers, and polybutadiene-based polyurethanes or their precursors, or mixtures thereof; with the proviso that the total amount of polybutadiene and unsaturated compounds is equal or less than 60 % by weight. The formulation is prepared via visbreaking of the above listed compounds with radical-generating agents.

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DECLARATION — Utility or Design Patent Application

POWER OF ATTORNEY As a named inventor, I hereby appoint the following registered practitioner(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: Customer Number 000530

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

NAME OF SOLE OR FIRST INVENTOR: [ ] A petition has been filed for this unsigned inventor

Given Name (first and middle [if any]): Francesco | Family Name or Surname: Mascia

Inventor's Signature: X Francesco | Date: 3/8/02

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Mailing Address: Via Cavour, 1

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NAME OF SECOND INVENTOR: [ ] A petition has been filed for this unsigned inventor

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Inventor's Signature: X | Date

Residence: City: Bulgarograsso | State | Country: Italy | Citizenship: Italy

Mailing Address: Via ai Prati, 4

City: Bulgarograsso | State | ZIP: I-22070 | Country: Italy

[ ] Additional inventors are being named on the ____ supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto.

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NAME OF SOLE OR FIRST INVENTOR: [ ] A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])	Francesco	Family Name or Surname	Mascia
Inventor's Signature	X	Date	
Residence: City	Guanzate	State	
Country	Italy	Citizenship	Italy
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City	Guanzate	State	
ZIP	I-22070	Country	Italy

NAME OF SECOND INVENTOR: [ ] A petition has been filed for this unsigned inventor

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Inventor's Signature	X Franca Arrighi	Date	5/21/02
Residence: City	Bulgarograsso	State	
Country	Italy	Citizenship	Italy
Mailing Address:	Via ai Prati, 4		
City	Bulgarograsso	State	
ZIP	I-22070	Country	Italy

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